Tuesday, July 28, 1998 CARBONACEOUS CHONDRITES I 8:30 a.m. Walton Theatre

Chairs: C. L. Duke M. K. Weisberg

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Noll K. Döbeli M. Grambole D. Herrmann F. Krähenbühl U.*

Fluorine Enrichment on the Surface of Antarctic C3O and H Chondrites by Nuclear Reaction Analysis (NRA) and the Sources of this Terrestrial Fluorine

AN EXPERIMENTAL STUDY OF HYDROUS MINERAL FORMATION REACTION BETWEEN ENSTATITE AND WATER VAPOR. N. Imae¹ and K. Shinoda², ¹Antarctic Meteorite Research Center, National Institute of Polar Research, 1-9-10 Kaga, Itabashi, Tokyo 173-8515, Japan, ²Department of Geosciences, Faculty of Science, Osaka City University, Sugimoto 3-3-138 Sumiyoshi, 558-8585, Japan.

According to the equilibrium condensation theory, it has been predicted that hydrous mineral formation reactions between silicates and water vapor occur in the primordial solar nebula [1]. On the other hand, it has been shown that this is inhibited from the reaction kinetics based on the collision theory in the primordial solar nebula [2]. However, no systematic experimental studies to form hydrous minerals by the reaction have been carried out. In the present study, we experimentally examined this reaction making use of the newly designed experimental method.

Autoclave (HU-25) with inner capsule of teflon was used for experiments. Starting material of enstatite single crystal (~1.5 mm) was put into the prepared inner teflon capsule. Ion exchanged water (a few cm³) was put into the outer usual teflon capsule. Thus direct contact between enstatite and water was escaped. Enstatite single crystals were synthesized using the flux method [3]. When this assembly is heated in a controlled electric furnace less than the critical point of water (374.15°C), water vapor in equilibrium with water liquid is formed around the enstatite and this is the environment for the enstatite to be able to react with water vapor. Since this condition is in the range of hydrous minerals stability field, hydrous mineral formation reaction might occur.

Temperature was controlled to be 230°C, which is upper limit of the teflon. The pressure is estimated to be 22 atm. Run duration was two months. After the heating, the change of the surface was not observed under the optical microscope. (In preliminary studies using natural pyroxenes, reaction rim was not observed from the thin sections under optical microscope and scanning electron microscope.) But infrared (IR) absorption spectra of enstatite before heating and after heating were measured by Fourier transform infrared microspectrometer (Shimadzu FTIR 4200 and IR microscope IMS-1 with wire grid polarizer). As a result, a sharp absorption peak was observed at 3692 cm⁻¹ for the enstatite after heating suggesting the formation of hydrous mineral.

Expected reaction is that

 $4MgSiO_3(Enstatite) + 2H_2O(g) = \\ Mg_3Si_4O_{10}(OH)_2(Talc) + Mg(OH)_2(Brucite) \ or$

 $6MgSiO_3(Enstatite) + 3H_2O(g) = \\ Mg_3Si_2O_5(OH)_4(Antigorite) + \\ Mg_3Si_4O_{10}(OH)_2(Talc).$

Both reactions forms two hydrous phases, but only single peal can be observed. The spectra to determine the phase will be analyzed in detail.

Thus we succeeded in the formation of hydrous mineral formation between enstatite and water vapor. The phase and the probable reaction equation are necessary to determine. The rate of the hydrous mineral formation between enstatite and water vapor expected from the present experimental study will be much smaller than that from the theoretical estimation by [2]. This might imply that silicate anhydrous minerals are quite difficult to react with water vapor even if the condition is the water vapor enriched compared with the solar nebula. Therefore negligibly small amount of hydrous minerals in chondrites would have formed by hydration in the primordial solar nebula.

Acknowledgments: Synthesized enstatite single crystals were kindly supplied by Mr. S. Tachibana in Osaka University. I am grateful to Dr. A. Tsuchiyama in Osaka University for his suggestion of using autoclave for the hydration experiments. This work is supported by Grant-in-Aid for Scientific Research from the Ministry of Education of Japan (No. 09740402).

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EXPERIMENTAL SIMULATION OF FAYALITIC RIMS ON OLIVINE: KINETIC CONSTRAINTS. R.

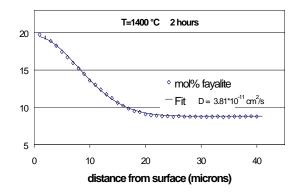
Dohmen, S. Chakraborty, H. Palme, and W. Rammensee, Institut für Mineralogie und Geochemie, Universität zu Köln, Zülpicher Str. 49 b, 50674 Köln, Germany, (dohmenr@min.uni-koeln.de).

Introduction: In Allende and other CV-chondrites forsteritic olivine is often associated with FeO-rich olivine. These fayalitic olivines occur either as up to 50 μ m wide rims with rather uniform composition around forsteritic cores and/or as zoned olivines with decreasing fayalite contents towards the core. These apparent diffusion profiles have variable lengths, from a few μ m to 50 μ m (e.g., [1], [2]). The maximum FeO-contents in both settings ranges from 20 to 45 mol% FeO. The variabilities in olivine composition and in diffusion lengths of FeO-profiles reflect a variety of formation conditions.

Earlier high temperature (1400 °C) Knudsen-cell mass spectrometry experiments [3] have shown that forsterite incorporates gaseous Fe (from Fe-metal) while Mg is released to the gas phase. In experiments with single crystals of San Carlos olivine 200 μm thick rims with high fayalite contents (>90 mol%) were observed, whereas, in experiments with small polycrystals of forsterite, but otherwise identical conditions, much lower contents of FeO in olivine were found (6 mol%). Further experiments with single crystals of SC olivine were made to better understand formation conditions of FeO-rich olivines.

Experiments: Preliminary results show that the run products and the reaction paths are strongly dependent on the exposed olivine surface area. A large surface area leads to a classical Fe/Mg exchange diffusion profile (Fig. 1) in olivine, whereas in experiments with a small surface area broad FeO-rich fayalitic rims are produced [3]. The observed diffusion profiles of Fe can be modeled with available diffusion data [4].

Figure 1:



Interpretation: A small exposed olivine surface leads to low Mg-partial pressures within the Knudsen cell, because the effusion rate of Mg-vapor through the hole of the lid of the Knudsen cell exceeds the evaporation rate of olivine. The Fe-metal buffered Fe-partial pressure is, however, not affected by the exposed olivine surface. The higher the Fe/Mg ratio in the gas the more fayalitic is the condensed olivine. In the extereme case, very high FeO/MgO-ratios will lead to the formation of an FeO-rich melt on the olivine surface eventually crystallizing as a broad fayalitic rim.

Implications: The experiments show the drastic influence of kinetics in processes leading to fayalite-rich olivine formation by reaction with a gas phase. The favalitic olivine formation is largely controlled by variable Fe/Mg ratios in the gas phase, which result from the difference in kinetics of evaporation of Femetal (fast) [5] vs. forsterite (slow) [6]. In the past, we have attributed fayalitic rim formation in Allende to oxidizing conditions in the nebula. The new results demonstrate that the kind and extent of favalitic olivine formation depends sensitively on details of the oxidizing event. For example, even a short heating event is capable producing broad favalitic rims. Thus, the variety of fayalitic rims and FeO-zoning observed in Allende olivine can be produced by locally different conditions and durations of heating during a single oxidizing event.

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Q-NITROGEN IN CO3 CHONDRITES. K. Hashizume^{1,2} and Y. T. Nakaoka¹, ¹Department of Earth and Space Science, Osaka University, Toyonaka, Osaka 560-0043, Japan, ²Centre de Recherche Pétrographique et Géochimique-CNRS, B.P.20, 54501 Vandœuvre-lès-Nancy Cedex, France (kohash@crpg.cnrs-nancy.fr).

It is widely observed that isotopic ratios of N in bulk meteoritic samples are largely different among chemical groups [e.g., 1,2], or even among meteorites within the same group [3]. It is so various that we cannot infer from the bulk data the "normal" one, i.e., the $\delta^{15}N$ of the most common component in the solar system reflecting the one of the solar nebular gas and the Sun.

Most of the normal heavy noble gases (the P1 component), commonly observed among primitive chondrites, are known to be trapped in a phase named "Q," whose exact host phase is poorly known. It is insoluble by nonoxidizing acids, but is lost by oxidation. In this study, we analyzed the N isotopic composition correlated with the "Q" noble gases, using several Antarctic meteorites: Yamato 791717 (CO3), Y 82050 (CO3), and Y 793567 (L3). Yamato 791717, the chondrite studied in the furthest detail, contains only small or negligible amounts of presolar diamond and SiC, whose existence can be a disturbing factor for our study. A similar work has been done by [4] using Dhajala (H3.8).

We prepared acid residues mostly in accordance with the method described in [5]. We further performed colloidal separation and gravity separation to the acid residue of Y 791717. Finally, we treated part of the acid residue of Y 791717 with HClO₄ to confirm whether the N component we paid attention was lost by the treatment, one of the criterions a Q-component should meet. We extracted the gases using the stepwise combustion/pyrolysis method, and measured N, Ne, and Ar isotopic ratios and Kr and Xe amounts using a static-type mass spectrometer [6].

Yields of the acid residues ranged from 0.36 to 0.70 wt%. In the case of Y 791717, the acid residue lost 31%, 31%, and 87% of C, N, and 36Ar of its bulk sample respectively. We observed among the acid residue separates a N component with δ^{15} N values around -45 ~ -65%, which are mostly released at 650°-750°C by the combustion method, where release peaks of ³⁶Ar, ⁸⁴Kr, and ¹³²Xe were observed, and at >1200°C by the pyrolysis method. The elemental ratios among Ar, Kr, and Xe were essentially the same with literature ratios for the P1 component [7]. We observed correlation between the δ¹⁵N values and ²²Ne_{A2}/¹⁴N ratio among the HF-HCl residue, colloidal fraction and density separates of Y 791717. The Ne-A2 component is considered to be trapped in presolar diamond, which contains a N component with $\delta^{15}N \sim$ -340% [8]. The cause of the ~20‰ variation of the mean $\delta^{15}N$ values among the separates can be explained by contribution of different proportion of presolar diamond into them. We estimate the $\delta^{15}N$ value of the N component in the Y 791717 acid residue to be $-50 \pm 5\%$, in a diamond-free basis. This component was completely removed by oxidation, together with the "normal" noble gases. We observed a similar $\delta^{15}N$ value in the acid residue of Y 82050 (CO3). We consider that the Q-N component in CO chondrites possesses a $\delta^{15}N$ value typically of -50%.

A N component as light as $\delta^{15}N \sim -50\%$ was not observed in Y 793567 (L3). We observed, instead, a N component with $\delta^{15}N$ ranging from -12 to -17%, almost the same values with the one observed in Dhajala [4]. It is currently not clear whether the $\delta^{15}N$ value of Q-N in UOCs is truly different from the one in CO chondrites, or if the difference is due to a contribution of non-Q components to either or both of the chondritic groups.

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LIGHT NOBLE GASES IN PHASE Q OF COLD BOKKEVELD (CM2): NEON-E AND A NEW ³He/⁴He RATIO IN Q CLOSE TO He-A. H. Busemann, H. Baur, and R. Wieler, ETH Zürich, Isotopengeologie NO C61, CH-9092 Zürich, Switzerland (busemann.erdw.ethz.ch).

Introduction: Elemental and isotopic ratios for the primordial noble gas component Q [1-3] were obtained from a HF/HCl-resistant residue of the CM2 chondrite Cold Bokkeveld by on-line stepwise etching [4] with HNO₃.

Noble gases in phase Q of an essentially uniform composition are thought to have been widespread in the early solar system during accretion of the planets [5]. Therefore, Q-gases may be isotopically similar to nebular gases present before the beginning of the solar nuclear processing. Thus, He-Q was used to deduce protosolar abundances of D (burnt completely to ³He by early solar nucleosynthesis), and ³He [6]. These data are fundamental to test the standard Big-Bang Nucleosynthesis theory [7].

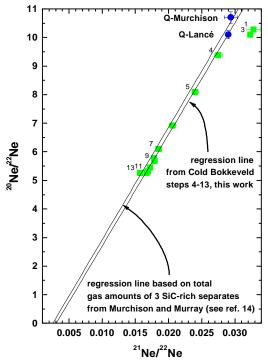


Fig. 1. Neon-E/Neon-Q mixing line from stepwise etching of Cold Bokkeveld.

Results: The gas release pattern indicates that no cosmogenic ³He and ²¹Ne affect the determination of Q-He and Q-Ne. This is because Cold Bokkeveld was exposed to cosmic rays for only 0.36 Ma [9], shorter than all other meteorites analysed by on-line etching. Similar to the CV3 chondrite Grosnaja [10], no He from other reservoirs with very different ³He/⁴He ratios could be identified either. The preliminary

 $(^3\text{He})^4\text{He})_Q$ deduced from the first 13 steps is $(1.42 \pm 0.01) \times 10^{-4}$, similar to Q-Grosnaja of $(1.45 \pm 0.01) \times 10^{-4}$, but significantly lower than the values obtained for Q-Allende $(1.59 \pm 0.04) \times 10^{-4})$ [4] and Q-Murchison 1.53×10^{-4} [2]. The new data show that these old values were compromised by - rather small-cosmogenic ^3He contributions. Surprisingly, He-Q is very similar to He-A with $(^3\text{He})^4\text{He}_A \approx 1.4 \times 10^{-4}$ [e.g., 2]. Despite this, He-Q must exist as an independent component, since Ne-Q and Ne-A are clearly distinct, and the ratios of He to any other noble gas are uniform in all steps. This new ratio is higher than the jovian ratio $(^3\text{He})^4\text{He} = (1.1 \pm 0.2) \times 10^{-4})$ [11] which is also used as the protosolar ratio to calculate protosolar D abundances [12].

The Ne-plot (Fig. 1) shows an almost perfect two-component-mixing between Ne-E and Ne-Q. No hints for accompanying gases (e.g., Xe-s [13]) could be found. The regression line is almost identical to that one defined by Zinner et al. [14] from SiC-rich samples of Murchison and Murray (CM2). This indicates that Cold Bokkeveld, Murchison and Murray contain a SiC population of very similar composition.

The elemental ratios of Q-Cold Bokkeveld are, in general, similar to those of other unequilibrated chondrites, although, Ne and, especially, He are somewhat more abundant than in other chondritic classes (as also seen in Murchison [2]), indicating less severe metamorphic losses on the CM parent body.

Acknowledgement: Work supported by the Swiss National Science Foundation.

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DISTRIBUTION OF MODERATELY VOLATILE ELEMENTS IN RENAZZO AND OTHER CR CHONDRITES. P. Kong¹, H. Palme¹, M. Ebihara², and G. Kurat³, ¹Institut für Mineralogie und Geochemie, Universität zu Köln, Zülpicherstrasse 49b, 50674 Köln, Germany, ²Department of Chemistry, Faculty of Science, Tokyo Metropolitan University, Hachioji, Tokyo 192-03, Japan, ³Naturhistorisches Museum, Postfach 417, A-1014 Vienna, Austria.

Chondrules, bulk metal, rim and matrix samples were isolated from the Renazzo meteorite and analyzed by INAA. A thin section of Renazzo was studied by EMPA. Bulk metals and non-magnetic fractions from four other CR chondrites (Acfer 209, Y 793495, PCA 91082 and EETA 92042) were also studied by INAA. Fine metal (< 0.5 mm) in CR chondrites is located mostly inside and coarse metal outside chondrules. Both types of metal exhibit similar compositional characteristics, suggesting a genetic relationship. Compared with Ni and Co, the moderately volatile siderophile elements Au, As, Sb and Ga are depleted in CR metal and the degree of depletion correlates with the volatility of the elements. In contrast, the non-magnetic fraction, primarily chondrule silicates and matrix, is enriched in these elements. The Renazzo matrix, although variable in Al and Ca, has essentially CI Zn/Sc and Se/Sc ratios. Na/Mg, K/Mg and Ga/Ni ratios in the Renazzo matrix are even higher than those of CI.

There are several possibilities for the fractionation of moderately volatile elements among components of CR chondrites: fractionation during condensation [1,2], evaporation during chondrule formation [3,4] and redistribution during aqueous alteration. The CR Acfer and Antarctic meteorites and Renazzo have experienced different degrees of aqueous alteration [5], but their metal compositions are very similar. In addition, it seems unlikely that the degree of hydrothermal leaching is a function of volatility of the element. Obviously, the difference in volatile siderophile element pattern between metal and matrix of CR chondrites cannot be the result of redistribution during aqueous alteration.

If chondrule precursors and matrix formed in the same nebular region and the fractionation of moderately volatile elements occurred during condensation, the absolute concentrations of volatile siderophile elements will depend on the size of metal grains, but the elemental pattern should be the same, independent of grain size. The obvious difference in volatile siderophile element pattern between bulk metal and

matrix, therefore, is not the result of fractionation during condensation of chondrule precursors and matrix in the same nebular region.

Another possibility involving fractionation by condensation is formation of chondrule precursors and matrix in different nebular regions. Both the chondrule fraction and the matrix of Renazzo have Mgnormalized refractory lithophile element concentrations identical to CI, different from most of other chondritic meteorites [6]. In addition, the chondrules and the matrix of Renazzo have the CI Cr/Mg ratio, while OC, CV, and CO components have lower ratios. It seems unlikely that the rare group of CR meteorites happened to have sampled chondrule and matrix materials both having CI refractory lithophile elements/Mg and Cr/Mg ratios from different nebular regions.

We, therefore, conclude that the fractionation of moderately volatile siderophile elements between metal and matrix of CR chondrites resulted from evaporation during formation of chondrules from matrix. The elevated Na, K and Ga concentrations in the Renazzo matrix may be best explained as partial recondensation of these elements onto the matrix. Although, the CI Se/Sc and Zn/Sc ratios of the Renazzo matrix and the low ratios of Renazzo chondrules are in agreement with the two component model proposed by Larimer and Anders [4], the gradual depletion of moderately volatile elements with increasing volatility in CR chondrites is most likely the result of evaporation followed by partial recondensation.

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THE BENCUBBINITE (B) **GROUP OF THE CR CLAN.** M. K. Weisberg¹, M. Prinz¹, R. N. Clayton², T. K. Mayeda², N. Sugiura³, and S. Zashu³, ¹American Museum of Natural History, New York NY 10024, USA, ²Enrico Fermi Institute, University of Chicago, Chicago IL 60637, USA, ³University of Tokyo, Tokyo, Japan.

The new bencubbinite (B) group is a quartet of metalsilicate chondritic meteorites that are members of the CR clan The quartet consists of Bencubbin, Weatherford, Hammadah al Hamra 237 (HH 237) and GRO 95551. Their silicates are mainly fragmented or complete chondrules of a single bulk composition, with textures ranging from barred olivine to cryptocrystalline. The distinguishing characteristics of Bencubbin [2-11] and Weatherford [9,12,13] are: (1) highly reduced silicates (Fs 3, Fa 3) (2) high metal abundances (50 vol.%), (3) chondrules of barred olivine composition, (4) FeNi with an ~solar Ni/Co ratio. (5) Mg-normalized refractory lithophile elements ~1xCI, highly depleted volatile lithophiles (<0.2xCI), (6) whole-rock oxygen isotopic compositions near the CR chondrite mixing line (Figure) and (7) ¹⁵N anomalies with ¹⁵N up to 1000‰. Ordinary, carbonaceous and R chondrite clasts are present in Bencubbin and Weatherford [3,6,11,13]. Two recent bencubbinite finds are HH 237, from the Sahara [14] and GRO 95551, from Antarctica [15]. Here, we present a petrologic and oxygen isotope study of HH 237 and establish the bencubbinite (B chondrite) group. N isotopic measurements of HH 237 are in progress. GRO 95551 has not been allocated yet.

Texturally, HH 237 is a mechanical mixture of silicate and metal fragments that are similar to Bencubbin but smaller. The silicates are generally mm-sized, mainly barred olivine to cryptocrystalline-textured chondrules, ranging from perfect spheroids to angular fragments. compositionally, they are similar to the cm-sized chondrule fragments characteristic of Bencubbin and Weatherford. Metal is present in two textural types. One is relatively large (up to 5mm) and near-spherical. The other, the more common type, is angular and mm-sized, similar to the silicate fragments. A silicate glass, with tiny (<1µm) immiscible blebs of FeNi, is present between the metal and silicate fragments. Similar material is found in Bencubbin and Weatherford, and was called "spontaneous fusion" texture, formed by impact melting [5]. Modally, HH 237 has (vol.%) 61 FeNi, 0.3 FeS, 14.3 olivine, 14.2 low-Ca pyroxene, 2.8 feldspar, and 7.2 other silicates. Olivine is mostly Fa 3 and Low-Ca pyroxene is Fs 3. Metal ranges (wt.%) from 4.5-13.0 Ni, 0.2-0.5 Co, 0.05-0.6 Cr, 0.05-0.5 P. Ni and Co are strongly correlated, with an ~solar Troilite abundance is low, and occurs as Ni/Co ratio. micrometer-sized blebs within some metal grains, mainly the large spheres; it contains 2.3-3.4 Cr, as in Bencubbin. CAIs are found in HH 237, but were not found in Bencubbin or Weatherford. The CAIs found are (1) a MgAl spinel-rich inclusion consisting of a cluster of 20-30µm spinel crystals, TiAl-rich Ca-pyroxene and melilite, and (2) a TiAl-rich Capyroxene-rich inclusion with forsterite and MgAl spinel. A hibonite-bearing inclusion was also reported [14]. Oxygen isotopes of Bencubbin and Weatherford are close to, but about 0.5% below, the CR chondrite line (Figure). HH 237 is similar, but has the heaviest oxygen composition of the bencubbinites, about 2% higher in ¹⁸O than in Bencubbin.

HH 237 is a new bencubbinite and the preliminary descriptions of GRO 95551 [15] shows that it too is a B chondrite. The presence of complete chondrules and CAIs in HH 237 strengthens the conclusion of [10] that bencubbinites are metal-rich chondrites. The reduced silicate compositions, metal compositions, refractory lithophile abundances and oxygen and nitrogen isotopic compositions indicate a strong relationship to CR and CH chondrites. All of these chondrite groups are members of the CR clan, as discussed in [1]. As in B chondrites, the CH chondrites are dominated by one chondrule type. The relatively small size of the chondrules in HH 237, and the presence of CAIs, suggest more of a continuum in the characteristics of the B and CH chondrites. In spite of diverse textures, chondrule sizes, presence or absence of hydrous components, and metal/silicate ratios, the CR clan members are kindred, with distinct characteristics.

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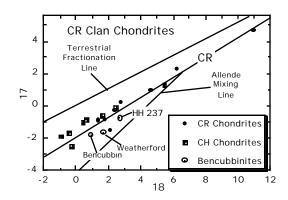


Figure. Whole-chondrite oxygen isotopic composition of CR, CH and B chondrites.

EXPERIMENTAL AQUEOUS ALTERATION OF ALLENDE (CV3). C. L. Duke and A. J. Brearley, Institute of Meteoritics, Department of Earth and Planetary Sciences, University of New Mexico, Albuquerque, NM 87131-1126, USA.

Introduction: A greater understanding of the reactions involved in the aqueous alteration of many carbonaceous chondrites can provide insights into the role of volatiles both in the solar nebula and on the asteroidal parent bodies. Several aspects of the fundamental characteristics of alteration reactions such as rates, mechanisms, and the role of fluid composition and water:rock ratio are poorly constrained. In order to investigate the mechanisms and rates of the reactions involved in the aqueous alteration of carbonaceous chondrites we have commenced a series of low-temperature hydrothermal experiments. The preliminary results of these experiments are presented below.

Methods: Small (~3mm) cubes of the CV3 chondrite Allende were heated with deionized water in Teflon lined Parr bombs. Experimental variables included temperature (100°C to 200°C), duration of heating (7 days to 60 days), and initial water: rock (W:R) ratio (1:1 to 6:1). After the experimental heating any residual water was analyzed using inductively coupled plasma-mass spectrometry (ICP-MS). Examination of the altered samples was carried out using a JEOL 5800LV SEM operated in low vacuum mode. Later the samples were carbon coated to analyze quantitatively the composition of the crystalline phases using EDS on the same SEM.

Results: In all cases, the samples had developed a reddish-brown discoloration after the experiment. Several new phases have crystallized on the surface of the cubes, the composition and morphology of which seems to be quite dependent on the experimental conditions. These new phases include Ca, Mg, and Na sulfates, Ca carbonates, and Fe-(hydro)oxides with a variety of different morphologies. The dominant sulfate crystals are hexagonal prisms of CaSO₄ that are perfectly formed in the lower W:R experiments and display considerable twinning and parallel growth as the W:R ratio is increased. Calcium carbonates also appear in a range of complexly twinned morphologies. Only in the highest W:R ratio experiment (6:1) did sulfate and carbonate phases coexist. Two morpho-

logically distinct Fe-(hydro)oxide phases are present on some samples. One is very finely crystalline and the other occurs as sub-micron spherules both in clusters and as individual grains. Preliminary SEM analysis of sample thin sections indicates that there has been some dissolution of the matrix olivine as well as oxidation of sulfide grains

Discussion: Our preliminary results demonstrate that even at low temperatures and after short reaction times quite extensive alteration of Allende has occurred. Oxidation of sulfides may provide S for the growth of sulfate phases, and also the Fe seen in the secondary Fe-(hydro)oxide phases. Matrix olivine may be the source of the Mg and some of the Fe in the experimental system. The source of Ca is not clear, although in one sample evidence of melilite breakdown has been observed. Other possible sources include high Ca pyroxenes in the matrix as well as the little remaining chondrule glass.

The secondary mineral assemblages found in our experiments bear a close resemblance to those found in CI chondrites, but differ from CM chondrites. Although calcite is abundant in CM chondrites [1], sulfates are rare, whereas in CI chondrites sulfate phases such as those that occur in our experiments are common [2]. We have not yet fully characterized the red-brown Fe-(hydro)oxide in our samples, but it may well be ferrihydrite, a phase that is also common in CI chondrites [3]. In addition, the spherular Fe-(hydro)oxide phase found in our experiments closely resembles the magnetite crystals within the framboids in CI chondrites [4].

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AMOEBOID OLIVINE INCLUSIONS AS INDICATORS OF HYDROTHERMAL ALTERATION OF CO3 CHONDRITES. L. J. Chizmadia¹, A. E. Rubin², and J. T. Wasson^{1,2,3}, ¹Department of Earth and Space Sciences; ²Institute of Geophysics and Planetary Physics; ³Department of Chemistry and Biochemistry; University of California, Los Angeles CA 90095, USA.

Amoeboid olivine inclusions (AOI) are fine-grained, irregularly shaped objects, typically 50–400 µm in size, that constitute several volume percent of carbonaceous chondrite groups other than CI. Although two varieties were previously dubbed "rimmed" and "unrimmed," the "rims" are actually alteration zones composed of massive olivine and are not restricted to the AOI surface. Because CO3 chondrites form a well-defined metamorphic/alteration sequence ranging from petrologic subtype 3.0 to 3.7 [1–3], studies of their AOI may offer insights into hydrothermal alteration. A qualitative measure of alteration is that the percentage of "rimmed" AOI tends to increase throughout the metamorphic sequence [3].

ALHA77307: AOI are minimally altered. They contain forsteritic olivine surrounding small patches of Al₂O₃-bearing augite or a fine-grained intergrowth of Ca-pyroxene and anorthite; augite (or Cpx+An) is more abundant in the interior of the AOI than near the surface. Opaque phases include chromite, kamacite and taenite.

Kainsaz: Most AOI show signs of incipient alteration; they consist of forsteritic olivine grain fragments (Fa₂₋₃ with <0.04 wt.% MnO) surrounded and cross-cut by thin (0.3–1.3 μ m) layers of more ferroan olivine (Fa_{≥15}). Opaque grains are tiny and very rare. Some AOI contain small veins of nepheline and relatively coarse inclusions of anorthite; ferroan olivine veins constitute ~20 vol.% of the AOI.

Warrenton: AOI are heavily altered. They tend to have lobate, amoeboid shapes and consist of ~10-µm-thick layers of ferroan olivine (Fa₃₀₋₃₅ with ~0.2 wt.% MnO; [1]) surrounding and cutting through interior regions consisting of augite partially surrounding Ca-plagioclase; the ferroan olivine layers constitute 50-75 vol.% of the AOI.

Recent observations indicate that CV3 chondrites experienced hydrothermal alteration [4], with the oxidized CV subgroups being more altered (and containing more "rimmed" AOI [3]) than the reduced subgroup. The same

trend may hold for CO chondrites wherein the most reduced, metallic-Fe-Ni-rich members (i.e., Kainsaz, Colony, Y-81020) are among the least altered. However, ALHA 77307, which is even less altered (but deviates from CO chondrite compositions [5]), contains relatively little metallic Fe-Ni [6].

Amoeboid olivine inclusions also occur in CR chondrites. These rocks have experienced significant aqueous alteration, but only moderate heating. Some of the CR AOI consist of relatively thick (10–40 μm) forsteritic olivine layers (Fa<1.5) surrounding and cutting through interior regions consisting of Ca-pyroxene partially surrounding Caplagioclase [7]. The texture and mineralogy of these AOI are somewhat similar to those in Warrenton. The main difference is that the AOI olivine grains in Warrenton are more ferroan, an apparent reflection of diffusive exchange with FeO-rich matrix material. It seems plausible that fluid alteration with only moderate heating is sufficient for AOI to produce their olivine "rims."

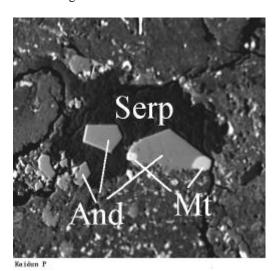
Our studies of AOI are aimed at (a) characterizing unaltered AOI (as in ALHA 77307) and examining the possibility that these objects are the earliest-formed chondrules [8], (b) documenting the beginnings of the AOI alteration process, and (c) understanding the mineralogical and textural transformations occurring throughout the metamorphic sequence.

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THE KAIDUN METEORITE: POSTACCRETIONARY ANDRADITE-MAGNETITE-SERPENTINE PRECIPITATION. F. Brandstätter¹, A. Ivanov², and G. Kurat¹, ¹Naturhistorisches Museum, Postfach 417, A-1014 Vienna, ²Vernadsky Institute of Geochemistry and Analytical Chemistry, 117334 Moscow, Russia.

Introduction: Garnets are rare in meteorites but at least four members of the garnet group are know to occur [1, 2]. Most of them were described as accessory minerals in carbonaceous chondrites. A few garnets were found to be products of high pressure shock events in ordinary chondrites. Here we report on the occurrence of euhedral andradite crystals in pockets of section #01.3.d1 from the Kaidun meteorite, a polymict breccia containing various clasts of oxidized and reduced lithologies [3].

Results: Perfectly shaped andradite crystals (up to 100 μ m across) are embedded in a matrix of serpentine (Serp; Fig.). Andradite (And) protrudes from the walls of the cavities towards their interior. Some andradites are intergrown with subhedral magnetite (Mt; up to 20 μ m across). In places, isolated grains of magnetite occur within the serpentine groundmass which apparently fills irregularly shaped pockets in the CM and CI lithologies of Kaidun.



The chemical composition of andradite, magnetite and serpentine is rather uniform. Andradite has an average composition (wt%) of: 35.5 SiO₂, <0.02 TiO₂, 1.03 Al₂O₃, 0.03 MnO, 27.0 FeO, 0.11 MgO, 31.2 CaO. The composition of magnetite is close to that of pure Fe₃O₄ except for an unusually high SiO₂ content (90.0 FeO, 2.2 SiO₂, <0.02 TiO₂, 0.1 Al₂O₃, 0.8 MgO, 0.4 CaO). Serpentine consists of fine-grained intergrowths of sub-micrometer-sized grains with an average chemical composition of: 37.3 SiO₂, 2.6 Al₂O₃, 33.6 MgO, 6.3 FeO (wt%).

Discussion: Since the first report of meteoritic andradite [4] this (Ca, Fe3+)-garnet has been described mainly in association with refractory inclu-

sions in CV3 chondrites ([5]-[8]) and dark inclusions [9]. The formation of andradite was explained: by condensation on a pre-existing Type A inclusion [5], by formation in an oxidizing solar gas before capture inside a Type A CAI [7], by anhydrous alteration of chondrules via alkali-Ca exchange reactions [8], from CAIs by exchange reactions in the solar nebula under oxidizing conditions [9], and by water-rock interaction in an asteroidal environment [10].

Andradite has been found before in Kaidun [11], associated with pentlandite and pyrrhotite forming complex aggregates in the phyllosilicate matrix of a CM lithology. The andradite bearing pockets described in our study exhibit some unusual features: (i) the euhedral shape and the direction of growth of the garnet crystals towards the interior of the pockets, (ii) the chemical homogeneity, (iii) the texture and chemical homogeneity of the serpentine groundmass. Therefore, we believe that andradite and magnetite were precipitated into pre-existing cavities and/or veins. Fluids or liquids rich in Ca and Fe were locally mobilized under highly oxidizing conditions. Redeposition in open cavities enabled undisturbed growth of andradite. From terrestrial environments it is known that andradite can form at low temperatures in skarns, in hydrothermal veins[12], and even by fumarolic activity[13]. As indicated by the textural relationship, magnetite either co-precipitated with or was formed before andradite. A late stage process at relative low temperatures filled the cavities with serpen-

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AQUEOUS ALTERATION ON THE CI PARENT BODY: EVIDENCE FROM OXYGEN AND CARBON ISOTOPIC STUDIES OF SINGLE CARBONATE GRAINS FROM ORGUEIL. K. L. Zito¹, K. D. McKeegan¹, J. F. Kerridge², I. D. Hutcheon³, and L. A. Leshin¹, ¹Department of Earth and Space Sciences, University of California, Los Angeles, CA 90095-1567, ²Department of Chemistry, University of California at San Diego, La Jolla, CA 92093, 3Isotope Science Division, Lawrence Livermore National Laboratory, Livermore, CA 94551.

Carbonate minerals in CI chondrites formed by aqueous alteration processes early in the history of the CI parent asteroid [1,2]. Stable isotopic studies of "bulk" CI carbonates [e.g., 3] have demonstrated variations in $\delta^{13}C_{\tiny PDB}$ and $\delta^{18}O_{\tiny SMOW}$ which imply a complex history of the CI asteroid, possibly including the formation of multiple carbonate generations. To examine this history in more detail, we determined $\delta^{18}O$ and $\delta^{13}C$ values in 64 spots in 29 carbonate grains (5 calcite, 7 dolomite, and 17 breunnerite) from Orgueil using the UCLA ims1270 ion microprobe.

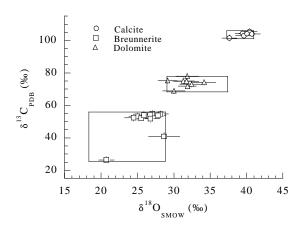
Dolomite and breunnerite grains were obtained from heavy mineral separates, potted in epoxy, and polished; calcites were analyzed in thin section USNM 234-2 (described by [4]). Some of the ⁵³Cr breunnerites contain excess indicating ⁵³Mn [2]. crystallization with live Isotope measurements were made in 15-20 µm spots using published techniques [5]; overall accuracy is ~1-2‰ in δ^{18} O and δ^{13} C. Major element compositions of all analysis spots were determined by electron microprobe.

Individual points in the Figure represent analyses where both $\delta^{13}C$ and $\delta^{18}O$ were determined in the same spot or region; the boxes encompass the range of all analyses (including cases where data were obtained for only C or O). Each mineral type is characterized by a well defined field on the plot of $\delta^{13}C$ vs. $\delta^{18}O$ with isotopic values increasing from breunnerite (+26.4 $< \delta^{13}C < +56.4\%$; +18.3 $< \delta^{18}O < 28.7\%$) to dolomite (+68.9 $< \delta^{13}C < +77.9\%$; +29.1 $< \delta^{18}O < +37.4\%$) to calcite (+101.5 $< \delta^{13}C < +105.6\%$; +37.0 $< \delta^{18}O < +40.9\%$). No appreciable isotopic variation within individual grains was observed, however, some variability among grains of similar mineral type exists, e.g., two breunnerites with high FeO contents exhibit distinct isotopic values.

The magnitude of the isotopic variations in the Orgueil carbonates, especially in $\delta^{13}C$, is too great to result from temperature variations in the fluid from which the carbonates precipitated. Thus, the coupled variation of the oxygen and carbon isotopic compositions implies that different Orgueil carbonate minerals formed during mixing of two isotopically distinct reservoirs. The ~22 ‰ variation in $\delta^{18}O$ could be explained by progressive aqueous alteration of silicates on the CI asteroid. In this scenario, initially isotopically heavy fluids evolve during alteration of

the isotopically light CI protolith, consistent with previous suggestions of fluid evolution on the CI parent body [6,7]. This interpretation requires that calcites formed early, followed by dolomites, and finally breunnerites and is consistent with suggestions, based on ⁸⁷Rb-⁸⁶Sr and ⁵³Mn-⁵³Cr, that dolomite predates breunnerite [2,8].

The end member components responsible for the C isotope mixing are less clear. In particular, the identity of the high δ^{13} C end member is unknown. The isotopic composition of the low δ^{13} C end member could be consistent with a precursor to the organic material that is presently found in Orgueil [e.g., 9], but the process by which this originally reduced material was converted into CO₃²⁻ in the fluid is still The generally primitive (solar) and unresolved. constant composition of different bulk Orgueil aliquots suggests that the variability of cations in carbonate production reflects localized solubilization rather than large-scale transport, but the mobilization of volatile C and O components over longer distances cannot be ruled out.



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ORGANIC SYNTHESIS DURING AQUEOUS ALTERATION OF CARBONACEOUS CHONDRITES: COMPARISON OF CM AND CI GROUPS. M. Schulte, Department of Earth and Planetary Sciences, Washington University, St. Louis MO 63130 and NASA Ames Research Center, MS 239-4, Moffett Field CA 94035 (e-mail: mschulte@mail.arc.nasa.gov).

Introduction: Secondary processing of the carbonaceous chondrites is at least in part responsible for some component of the soluble organic compounds found in these meteorites. The well-documented aqueous alteration which occurred on the parent body(ies) of carbonaceous chondrites has been investigated mainly in the context of the most well-studied of these meteorites, Murchison, a CM group chondrite. Previous theoretical work [1] has demonstrated the link between the alteration of the mineral assemblages and production of organic compounds. The CI group, however, has experienced more extensive alteration than the CM group (2-4) and also contains abundant soluble organic material [5-6]. The purpose of this study is to explore the relationship between the extent of alteration of the CI group and the potential for synthesis of soluble organic material during the alteration. Model calculations for the CI group are compared to those for the CM group.

Modeling parent body processes: Despite the extensive nature of alteration products present, the CI chondrites are considered to be the most primitive meteorites because their bulk chemistry is identical to that of the solar photosphere [7-8]. For this reason, I adopted the average bulk composition of the CI chondrites from Dodd [9] for modeling the aqueous alteration processes using mass-transfer, reaction-path calculations. The nature of the fluid responsible for the alteration remains speculative, but I have chosen a relatively oxidizing, dilute solution saturated with CO₂, CO, CH₄, N₂ and NH₃ as the starting fluid. I minimized the Gibbs free energy of closed chemical systems using stable and metastable equilibrium constraints at various temperatures that may have been reached during the alteration process.

Results: The calculations result in mineral assemblages which are consistent with observations of CI chondrites. For example, at 100°C, the approach to equilibrium produces serpentines (antigorite, cronstedtite, greenalite), dolomite, carbonates, sulfides, chlorite and other phyllosilicates. This mineralogy is similar to that previously calculated for CM chondrites [1], except for the appearance of dolomite for the CI, in agreement with the presence of this mineral in CI and its absence in CM chondrites. The final alteration event in CI chondrites may have involved a more severe loss of volatiles at higher temperatures

than experienced by the CM group. This is supported by the presence of sulfate veins which cross-cut previously formed alteration.

Another similarity to the CM models is the predicted appearance of soluble organic and the nature of these compounds. Carboxylic acids dominate the organic carbon speciation in the calculations, followed by amides, metal-organic compounds, ketones, alcohols, and other organic compound families.

The results of these calculations suggest that the nature of the alteration of the CI and CM chondrites was probably very similar, and that the presence of soluble organic compounds in each of these meteorite classes is due at least in part to formation from inorganic precursors during the aqueous alteration event(s) responsible for the observed mineral assemblages.

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RACEMIZATION OF METEORITIC AMINO ACIDS. B. A. Cohen¹ and C. F. Chyba, University of Arizona, Tucson AZ 85719 ¹(bcohen@lpl.arizona.edu).

Amino acids are the building blocks of proteins, among the most biologically critical molecules. It was first noted in the 1800s that the vast majority of amino acids used by living organisms are the left-handed enantiomers (laevorotatory, or L) rather than the right-handed enantiomers (dextrorotatory, or D). However, the question of why life chose the L- over the D-enantiomers remains an outstanding question. Delivery of chirally enhanced extraterrestrial organic material might have been one way to enhance the concentration of L-amino acids on the earth and to encourage the choice of L-enantiomers in life processes. Amino acids were first found in the carbonaceous chondrite Murchison [1], but were reported to have a D/L ratio of ~1. Recently L-enantiomeric excesses have been reported in the Murchison amino acids [2,3]. If a nonracemic mixture of amino acids were incorporated into an asteroidal parent body, the chiral signature might be transmitted to Earth through meteorite delivery. This work investigates how much racemization takes place throughout an extraterrestrial amino acid's lifetime.

Racemization rate data was obtained for nine different amino acids: alanine, aspartic acid, glutamic acid, isoleucine, leucine, phenylalanine, proline, serine, and valine. D/L ratios were determined after a number of thermal history scenarios based on meteorite evidence and asteroid thermal models [4,5]. The process can also be worked backward, beginning at time t=now with some D/L ratio and determining the initial ratio.

Cronin and Pizzarello [2] find L-enantiomeric excesses in Murchison amino acids that are not easily

racemized (α -methyl amino acids), but a racemic mixture of the α -hydrogen amino acids investigated. Engel and Macko [3] find L-enantiomeric excesses in the more easily racemized α -amino acids alanine (D/L= 0.5) and glutamic acid (D/L= 0.3), though these results are controversial. Back-modeling reported excesses shows that these observed ratios could be produced by a wide range of initial D/L ratios. Whether the mechanisms for imposing chirality on amino acids could produce such starting ratios is unclear.

For the bulk of their history before we find them, meteoritic amino acids experience cold and dry conditions that prevent racemization, but racemization can quickly take place in an aqueous environment. The degree of racemization is quite sensitive to a number of parameters that are difficult to constrain, including duration and temperature of the liquid phase. The final D/L ratio does not give unambiguous information about the path which produced it. These results support the idea that delivery of an initial chirality to the Earth in the form of extraterrestrial organic material is possible, since there are some scenarios in which 100% racemization of an initial $D/L \neq 1$ amino acid mixture need not occur.

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FLUORINE ENRICHMENT ON THE SURFACE OF ANTARCTIC C3O- AND H-CHONDRITES BY NUCLEAR REACTION ANALYSIS (NRA) AND THE SOURCES OF THIS TERRESTRIAL FLUORINE.

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Terrestrial enrichment of F on the surface of Antarctic meteorites was investigated by the PIGE-NRA technique (Proton induced gamma emission, a class of nuclear reaction analysis [1,2]) using the nuclear reactions $^{19}F(p,\alpha\gamma)^{16}O$ and $^{19}F(p,p'\gamma)^{19}F$ and proton energies of ~3 MeV. With this technique, the F-profiles of the H-chondrites ALHA 77294, TIL 82409, LEW 86015 and the C3O-chondrites ALHA 77003 and ALH 83108 were measured. The F enrichments obtained for the H-chondrites show an analogous sequence as revealed by earlier results which were obtained by a different approach [3,4]. The selected samples have terrestrial ages in the range of 10'000 to 110'000 years and weathering indexes from A to C. The measured enrichment of F shows only a poor correlation with the weathering index and none with the terrestrial age of the samples analyzed. For two C3O- and one H-chondrite from the Allan Hills, the concentration of F in the unaltered sample material and in the topmost surface layer of ~0.04mm depth are given in Table 1. The surplus of F in the surface layer, stemming from terrestrial contamination, is also shown.

TABLE 1. F-contents in the surface layer and in the volume of one H5- and two C3O-chondrites in 10^{-6} g/g.

| | Surface | Pristine | F-excess in |
|------------|---------|------------|---------------|
| Meteorite | F-conc. | F-content | surface layer |
| ALHA 77294 | 118±6 | 10±2.3 | 108±6.4 |
| ALHA 77003 | 92±5 | 21 ± 2.3 | 71±5.5 |
| ALH 83108 | 232±10 | 17±2.2 | 215±10 |

The values of Table 1 were obtained by the reaction $^{19}F(p,\alpha\gamma)^{16}O$. Measurements with $^{19}F(p,p'\gamma)^{19}F$ on the topmost surface layer of ALHA 77294 and ALHA 77003 gave 98 \pm 18 and 89 \pm 15 10^{-6} g/g, respectively. Since different spots were investigated and the layer's depth was ~0.05 mm in this case, the agreement is quite satisfying. Terrestrial contamination should mainly occur during the time span an Antarctic meteorite resided on the surface of the ice.

So, its enrichment of F is an indication of this surface residence duration. Based on the integral enrichment in the topmost surface-layer of the two C3O-chondrites analyzed and in relation to the enrichment function deduced for ordinary chondrites, we come up with maximum exposition duration for ALHA 77003 and ALH 83108. The exposure time span for the latter was definitely longer than for ALHA 77003. Furthermore, the diffusion profiles for the C3O-chondrites are rather steep in comparison to the ones found for H-chondrites [2].

There are several sources for the terrestrial enrichment of F on Antarctic meteorites conceivable [2]. The previous hypothesis, assuming sea-salt aerosols to be the main source [4], has become rather remote after recent research results [5]. Thus, besides continental dust and layers of volcanic ash emerging in blue-ice ablation zones [6,7], the constant volcanic exhalations of Mt. Erebus (Ross Island), with their exceptionally high concentration of F [8], are proposed to play an important role as a dominant source for the terrestrial F enriched in Antarctic meteorites. This view will be further discussed together with additional indications [2].

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